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Magnetism of amorphous Dy-Tb-Co-type films

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Abstract. In this work a systematic study of magnetic properties of amorphous Tb_xCo_{100-x} and Dy_xCo_{100-x} films is presented. In a composition range $15 < x < 40$ at. %, temperature dependencies of spontaneous magnetization in a temperature interval 5 – 300 K as well as low-temperature 5 K hysteresis loops in magnetic field up to ± 70 kOe have been obtained. We demonstrated, that the experimental results can be interpreted in a framework of a model considering a variable sperimagnetic cone angle of rare earth atoms up to 150°. A series of Tb-Dy-Co films with a composition close to Laves phase has been obtained. We demonstrated that the variation of rare earth content with Co content fixed does not lead to a systematic lowering of magnetic hysteresis.

1. Introduction

Intermetallic systems (TbDy)-(FeCo) including a well-known commercially available alloy Terfenol-D [1] continue to attract attention as a base for perspective magnetostrictive materials. At the same time, the interest of researchers is largely focused on the thin-film state of these magnetic materials [2-4]. In particular, this trend is associated with the development of composite materials with a high magnetoelectric effect [5, 6]. In case of thin films, however, a combination of high magnetostriction with relatively weak magnetic anisotropy, as is the case in Terfenol-D, is difficult to achieve. This is due to the pronounced tendency of intermetallic compounds to amorphization when they are transferred to the film state, for example, using ion sputtering [7]. In this state, the regularity of the crystal field as well as the effect of combination of Tb and Dy disappears. In addition, the fluctuations of the distances between rare-earth atoms, magnetic moments of which are ordered due to the RKKY interaction, may lead to frustrated magnetic structure. As a result, the so-called sperimagnetic ordering arises in (TbDy)-(FeCo) films [8]. It is described as a combination of a collinear magnetic structure in a 3d-magnetic subsystem and a fan-like spatial distribution of magnetic moments of heavy rare-earth elements. In this case, the resulting magnetic moments of the subsystems of the rare-earth and 3d-atoms are oriented antiparallel to each other. High local magnetic anisotropy and strong magnetic hysteresis are characteristics of the sperimagnetic state, which make such films less appropriate for practical applications. In this regard, there is a problem of a search for new possibilities of controlling the parameters of sperimagnetic structure in alloys containing heavy rare earth elements and iron group metals. This paper attempts to solve this issue for the amorphous Tb-Dy-Co system.



2. Samples and techniques

In this work we synthesized and studied 110 ± 10 nm thick $\text{Tb}_x\text{Co}_{100-x}$, $\text{Dy}_x\text{Co}_{100-x}$ and $(\text{Tb}_{100-x}\text{Dy}_x)\text{Co}_2$ films ($0 \leq x \leq 100$ at.%) deposited onto Corning glass substrates and covered with protective seed and top 3 nm thick Ti layers. Mosaic Tb-Co, Dy-Co and Tb-Dy-Co targets with variable relative area of sputtered metals were used. The deposition process took place in Ar atmosphere at an operating pressure of 10^{-3} mm Hg and at the presence of the technological magnetic field of 200 Oe applied parallel to the substrate's plane. The elemental composition of all samples was determined using the Nanohunter X-ray fluorescence spectrometer with an uncertainty of 0.1 at.%. The thickness was controlled using the Dektak-150 mechanical profilometer. Magnetic measurements were carried out using MPMS XL-7 device, which allowed us to vary temperature and intensity of an external magnetic field in range of 5-350 K and ± 70 kOe respectively.

3. Results and discussion

Temperature dependencies of magnetization $M(T)$ measured on $\text{Tb}_x\text{Co}_{100-x}$ and $\text{Dy}_x\text{Co}_{100-x}$ films of several compositions ($0 < x < 50$ at.%) are shown in figure 1. In general, they are similar and have features typical for this type of magnetic material, which is characterized by an amorphous state, ferrimagnetism of the subsystems of rare earth elements (R) and cobalt, hybridization of 3d and 6s electronic bands of cobalt and rare earth elements [9]. Firstly, the amorphous state allows for a continuous change of composition and magnetic properties, in contrast to the discrete properties change typical for crystalline intermetallic compounds of the same composition. Secondly, the difference in exchange interaction in the cobalt and rare-earth magnetic sublattices leads to the typical non-monotonic temperature dependences of magnetization. Lastly, the hybridization of the R and Co electronic bands has a negative effect on the magnetism of Co, reducing its effective atomic magnetic moment down to zero at $x > 50\%$. Thus, for the films of certain compositions, the magnetic compensation point (e.g. curve 7 in figure 1) or an absent of magnetic ordering at room temperature (curves 5, 10 in figure 1) can be observed.

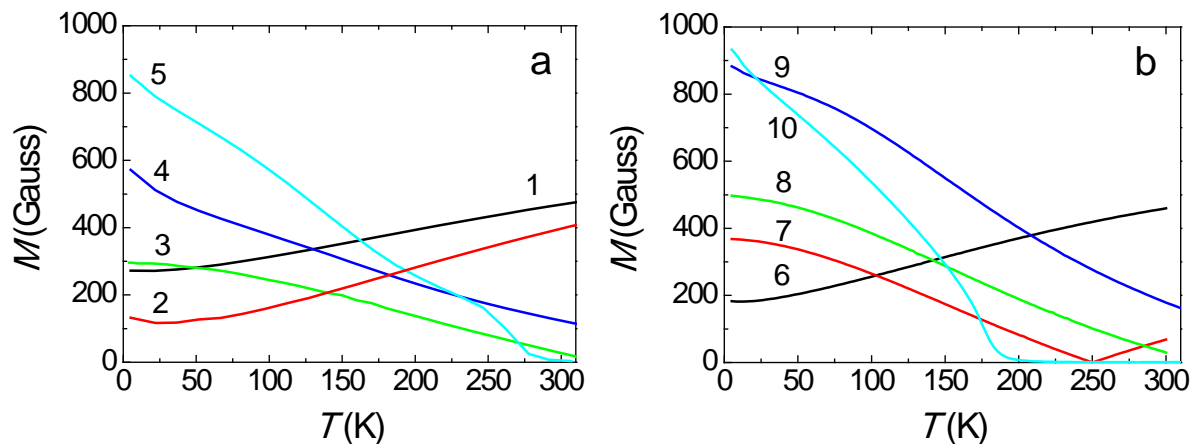


Figure 1. Temperature dependencies of magnetization of Tb-Co (a) and Dy-Co (b) films, measured in the magnetic field of 100 Oe for samples with different concentration of rare earth components: 1 – 8; 2 – 18; 3 – 22; 4 – 31; 5 – 43; 6 – 15,6; 7 – 24; 8 – 27; 9 – 27.5; 10 – 49 at.%.

Despite certain similarity of the electronic structure, Tb and Dy are known to have significantly different magnetic structure and magnetic anisotropy in both pure metal state and in intermetallic compounds [10]. Amorphous state, however, eliminates these differences. In particular, it can be seen in figure 2, which shows examples of typical low-temperature hysteresis loops measured on $\text{Tb}_x\text{Co}_{100-x}$ and $\text{Dy}_x\text{Co}_{100-x}$ films having R concentration range $15 < x < 40$. The main feature of the magnetization

reversal process, both at relatively small (figure 2a) and large (figure 2b) concentrations of R, is a significant change of magnetization in a high-field range.

Since the temperature was fixed at 5 K, such a character of $M(H)$ dependence cannot be related to the paraprocess. In fact, it is an indirect confirmation of the sperimagnetic nature of such films and reflects the change of the sperimagnetic fan angle in the external magnetic field. When the magnetic moment of the cobalt subsystem is dominating (figure 2a), an increase of H leads to the opening of the cone, otherwise (figure 2b) to its collapse.

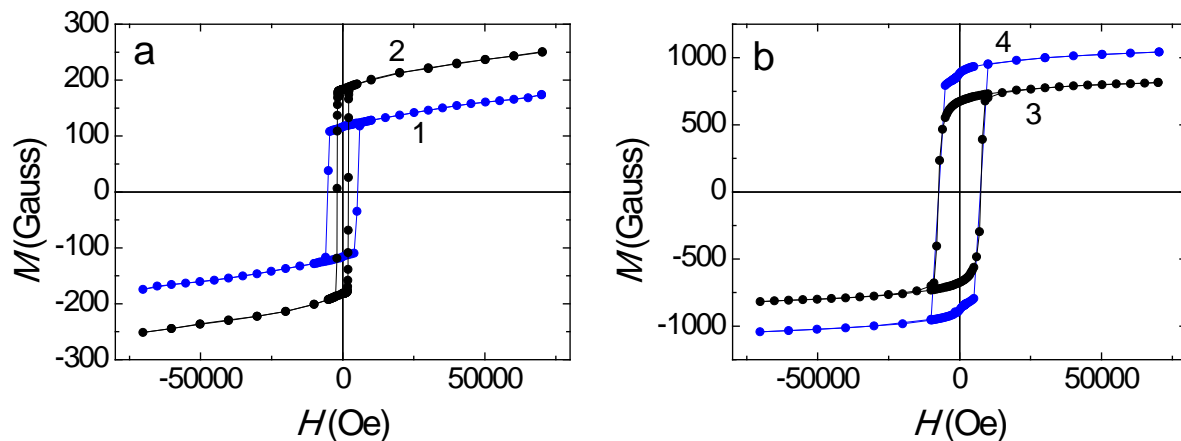


Figure 2. Hysteresis loops measured at 5 K on Tb-Co (curves 1, 3) and Dy-Co (curves 2, 4) samples with different concentrations of rare earth components: 1 – 18; 2 – 15.6; 3 – 35; 4 – 37.5 at.%.

In the framework of the model of collinear magnetic sublattices, it is possible to estimate average values of magnetic moments m_R per atom of the rare-earth elements. In this work, we used a concentration dependence of the average atomic magnetic moment of Co – m_{Co} estimated for amorphous La_xCo_{100-x} films (La does not carry a magnetic moment) [8]. The corresponding dependence $m_{Co}(x)$ is shown in figure 3a. As one can see, an increase in the concentration of the rare-earth element leads to a monotonic decrease in the average magnetic moment of Co atom until its complete disappearance at $x > 50$. Such dependence reflects the above-mentioned transformation of the electronic structure of Co in amorphous R-Co systems.

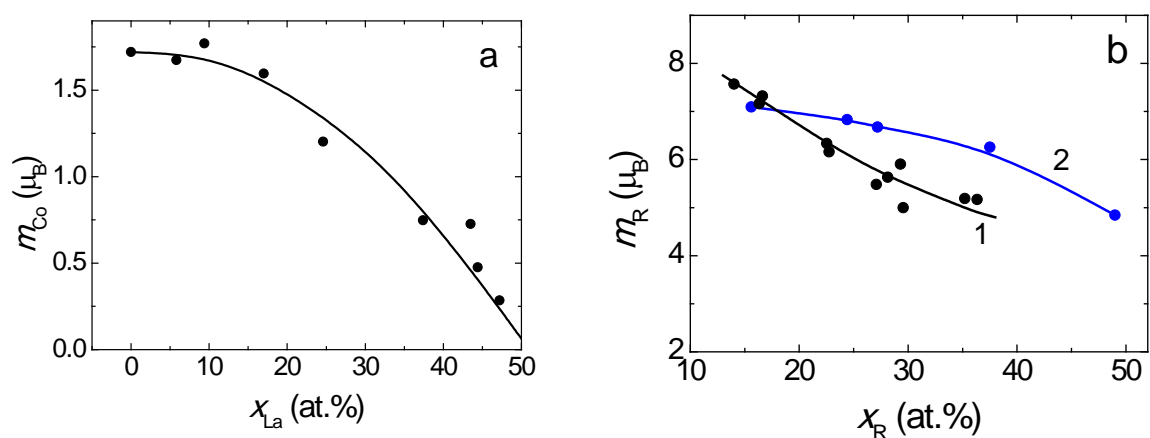


Figure 3. Dependencies of average atomic magnetic moments of Co (a), Tb (b, curve 1) и Dy (b, curve 2) on concentration of rare earth components in R_xCo_{100-x} films.

Figure 3b shows the concentration dependences of the average atomic magnetic moments of rare-earth elements. In both cases the level of experimental values of m_R is significantly lower than the values expected in free Tb and Dy atoms (9 and 10 μ_B respectively). Thus, we can conclude that the fanning cone opening angle of magnetic moments in R sublattices is very large, and according to estimates made under the assumption the uniformly filled fan, it reaches 150° . However, the course of $m_{Tb}(x)$ and $m_{Dy}(x)$ dependencies is somewhat different. It may reflect the same difference between rare-earth elements, which alters the magnetic properties of the crystalline counterparts.

According to the experimental results obtained on amorphous R_xCo_{100-x} films, the magnetic ordering at room temperature is maintained up to $x \sim 40$ at.%. This interval also includes the ratio between R and Co corresponding to Laves crystalline phases (RCo_2), on which the magnetostrictive material Terfenol-D is based. Figure 4 shows the dependencies of the coercivity H_c of amorphous films having composition close to the Laves phase but different concentration ratio between Dy and Tb. As one can see, the experimental points are scattered significantly. It does not allow to distinguish a certain tendency, but at the same time it shows a rather high overall level of magnetic hysteresis. Thus, unlike in crystalline counterparts, the magnetic anisotropy cannot be reduced in (Dy-Tb) Co_2 amorphous system by choosing a certain ration between concentrations of R elements. A possible solution could be to change the structural state of the films from amorphous to nanocrystalline by means of an appropriate heat treatment.

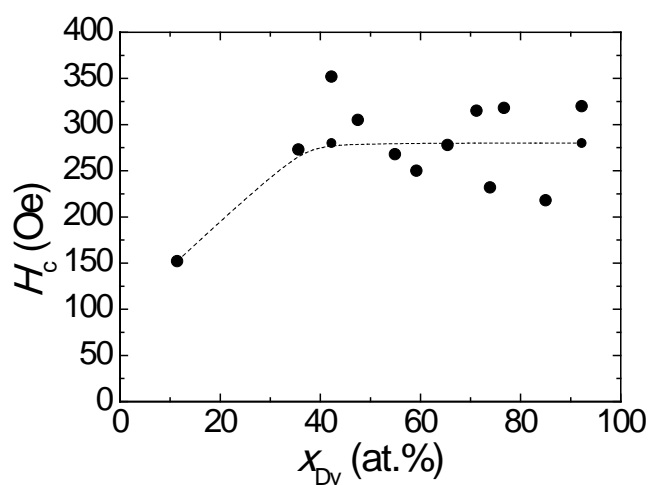


Figure 4. Dependency of coercivity on relative concentration of Tb and Dy obtained on $(Tb_{100-x}Dy_x)-Co$ films with a concentration of Co exceeding 65 at.%.

4. Conclusion

Magnetic properties of Tb_xCo_{100-x} and Dy_xCo_{100-x} films were systematically studied for $15 < x < 40$ composition range, at temperatures 5-300 K, and magnetic field up to 70 kOe. Both types of samples were confirmed to have a sperimagnetic ordering. The angle of sperimagnetic cone of rare earth atoms was shown to depend on films composition and can reach 150° . For Tb-Dy-Co films having composition close to Laves phases we did not observe a significant change of hysteresis due to the variation of a relative rare earth concentration.

Acknowledgement

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